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DETERMINATION OF NATURAL RADIOACTIVITY LEVELS IN SURFACE SOILS OF OLD PHOSPHATE MINE AT RUSSAIFA OF JORDAN

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ABSTRACT

The aim of this study was to evaluate the concentration levels of naturally occurring radioactive materials (NORMs) of ^{238}U and ^{232}Th series and ^{40}K in surface soil samples of old phosphate mine at Russaifa of Jordan. The activity concentrations of NORMs in soil samples were determined by means of gamma-ray spectrometry system using a hyper-pure germanium (HPGe) detector in a low background configuration. The ranges of specific activity concentrations (SACs) of ^{238}U, ^{232}Th and ^{40}K were found to be 27.55 ± 7.55 to 420.19 ± 60.25, 222.35 ± 8.15 to 366.68 ± 92.47 and BDL to 2.40 ± 0.20 Bq kg^{-1}, with mean values of 207.10 ± 21.02, 265.95 ± 30.84 and 0.895 ± 0.105 Bq kg^{-1}, respectively in the surface soil samples collected from old phosphate mine. The average values of 587.49 ± 179.6 Bq kg^{-1}, 264.53 ± 78.6 nGy h^{-1} and 0.32 ± 0.09 mSv y^{-1} for radium equivalent activities, absorbed dose rates in air outdoors and annual effective dose equivalent, respectively, while the average values of 1.58 ± 0.48 and 2.14 ± 0.84 for external and internal radiation hazard indices, respectively were calculated for the analyzed samples to assess the radiological hazards arising from natural radioactivity in surface soils. These obtained values in old phosphate mine were found to be higher than the internationally acceptable limits reported by UNSCEAR−1988, UNSCEAR−2000 and ICRP−1990.

KEYWORDS: Gamma-Ray Spectrometry, Hyper-Pure Germanium (HPGe) Detector, Specific Activity Concentration, Radium Equivalent Activity, Absorbed Dose Rate, Radiation Hazard Indices

INTRODUCTION

Radiation include external sources, such as cosmic rays and radioactive materials in the ground and in building materials, and internal sources resulting from inhalation and ingestion of naturally occurring radioactive materials (NORMs) in air and diet [UNSCEAR 1988, UNSCEAR 1993].

Radioactive materials are found naturally as trace elements in soil, rocks, building materials, ground water, air and vegetation. Radionuclides can be found in nature in three categories [UNSCEAR−1993]: firstly primordial radionuclides, such as ^{40}K and the radioisotopes from ^{238}U and ^{232}Th series and their products, which occur at trace levels in all ground formations. Primordial radionuclides, typically with half lives in the order of hundreds of millions of years. Secondly cosmogenic with long half lives, but the majority have shorter half lives than the primordial radionuclides, such as ^{3}H, ^{7}Be, ^{14}C and ^{22}Na, and thirdly human radionuclides with small amounts of activity compared to natural radioactivity due to the shorter half lives of many of the radionuclides [WHO−2004]. Activity concentrations of primordial radionuclides in rocks are usually in igneous rocks than sedimentary ones. Certain sedimentary rocks, notably some shales and phosphate rocks, are highly active, that contain of trace amounts of uranium (^{238}U) and thorium (^{232}Th) series [UNSCEAR 1988].

According to the Jordan Phosphate Mines Company (JPMC), more than 60% of the area of Jordan has phosphate. JPMC commenced its phosphate mining activities in 1935 in the Russaifa phosphate mine. In 1985 the JPMC closed the old phosphate mine at Russaifa, because its production has been reduced to low levels.

A significant contribution to natural exposure of humans is due to radon noble gas. Radon-222 (^{222}Rn) and ^{220}Rn are the gaseous radioactive products of the decay of the radium isotopes ^{226}Ra and ^{224}Ra, respectively, which are present in all terrestrial materials [UNSCEAR−2000]. The γ-emitting decay products of ^{222}Rn are lead (^{214}Pb) and bismuth (^{214}Bi) [5], which are found in radioactive secular equilibrium with radium (^{226}Ra) only if sealed to stop radon from escaping, while the decay products of ^{220}Rn are ^{212}Pb and ^{212}Bi, which are found in radioactive secular equilibrium with ^{224}Ra only if sealed to stop ^{220}Rn from escaping. From these decay products the estimated activity concentration in Bq kg^{-1} of ^{238}U and ^{232}Th can be obtained [5, 6].

A number of methods have been devised for determination of ^{238}U, ^{232}Th series and ^{40}K in environmental samples. Most widely used are inductively coupled plasma-mass spectrometry (ICP-MS) [7], X- rays fluorescence (XRF) [8], low level γ-ray spectrometry using NaI(Tl) detector [8], low-level liquid scintillation counting (LSC) [9] and high-resolution gamma-ray spectrometry system using a hyper-pure germanium (HPGe) detector [10 - 15]. The use of germanium detectors in high-resolution gamma-ray spectrometry is one of the most widely used procedures for the identification and quantification of unknown gamma-ray emitting radionuclides in environmental samples. It is a non-destructive technique, simplest analytical methods, rapidity and high sensitivity [12 - 15]. Applications of high-resolution gamma-ray spectrometry system have been used for determination of specific activity of ^{238U}, ^{232}Th series and ^{40}K in soil [11 - 16], building materials [17 - 19], phosphate rocks and phosphate fertilizers [20], water [21] and agricultural crops [10].

In the present work, high purity germanium (HPGe) detector-based gamma–ray spectrometer technique was used for identification and quantitative measurement of natural radioactivity levels of ^{238}U, ^{232}Th series and ^{40}K in surface soil samples collected from old phosphate mine at Russaifa in Jordan. Average radium equivalent activities, absorbed dose rate in air, annual effective dose equivalent, external and internal radiation hazard indices were estimated for the area under study. Comparison was made between the present experimental results and worldwide median values reported by UNSCEAR−1988, UNSCEAR−2000 and ICRP−1990 and published data by other workers [11 - 15 and 17].

EXPERIMENTAL PROCEDURE

Soil Sample Preparation

The surface soil samples were collected at 5 cm depth from the top surface layer from nine different locations across the old phosphate mine in Russaifa during 2013/2014, using clean polyethylene bags. Soil samples were properly marked and then sent to the lab to be processed. The stones were removed from the soil samples and were air dried in oven at 100^{o}C for two hours, and then crushed, ground to fine powder, passing through a 0.2 mm sieve, and homogenized. The mass of each soil sample was measured precisely. Marinelli beakers of 900 ml were filled with soil samples, closed by caps, and firmly sealed with plastic tapes to prevent the escape of radon (^{222}Rn and ^{220}Rn) from the Marinelli beakers. The sealed soil samples were stored for at least for one month or more prior to measurement in order to achieve radioactive secular equilibrium between ^{226}Ra, ^{232}Th and their short lived radon (^{222}Rn, ^{220}Rn), and their decay products, respectively [5].

Instrumentation

A hyper-pure germanium (HPGe) detector with high resolution gamma-ray spectroscopy low background counting system was used to determine the concentration levels of NORMs in soil samples. The spectrometry system consists of HPGe coaxial detector model number GMX30-76 EG & G Ortec, with active volume of 180 cm^3, operating voltage of – 4000 V, and the associated electronics, including preamplifier and amplifier were installed in a personal computer with 16384 channel multi channel analyzer (MCA) for data acquisition. The detector was surrounded by cylindrical lead shield of thickness 10 cm to reduce the background radiation with a movable cover. The lead shield contains an inner concentric cylinder of 0.2 cm thick copper to attenuate the X-rays stimulated in the lead shield itself. The Ortec software Gamma-Vision 32 was used for data analysis. Radioactive gamma reference sources of ^{109}Cd, ^{137}Cs, ^{57}Co, ^{60}Co, ^{54}Mn, ^{22}Na and ^{65}Zn from Spectrum Technique Inc. USA were used for calibration. The energy calibration was carried out by using multi radioactive standard sources emitting γ-rays of precisely known energy, and then identifying the peak position in channels with this energy. The efficiency calibration of the HPGe detector was carried out over the entire energy range of interest using standard radioactive sources. The energy resolution (FWHM) of HPGe detector at 1.33 MeV from ^{60}Co is 1.9 keV, and the relative efficiency at 1.33 MeV, ^{60}Co is 30%.

Spectrum for each sample was collected for 79200 seconds counting time to reduce statistical errors. The area under each identified peak of the spectra was calculated individually. The background carefully measured by filling the Marinelli beaker with inactive de-ionized water for the same counting time 79200 s, then placed on the detector to get the same geometry. The net number of counts for each photo peak was obtained by subtracting the background counts from the total counts in the same photo peak, for 79200 seconds.

Determination of NORMS Activity Concentration

The activity concentrations of unknown γ-ray emitting radionuclides in environmental samples have been determined by means of a gamma-ray spectrometry system using a hyper-pure germanium detector in a low background configuration. The majority of NORMs belong to the radionuclides in the ^{238}U and ^{232}Th series, and the single decay radionuclide, ^{40}K [IAEA-2003]. In the ^{238}U and ^{232}Th decay series, radon (^{222}Rn and ^{220}Rn) are the only radionuclides found in the gaseous state and they can emanate naturally from soil and rocks.

The area under each photopeak of the spectrum was calculated individually and was subtracted from the background radiation. The specific activity concentrations of individual radionuclides *i* for photo peak at energy *E* in soil samples can be calculated by using the following expression [16 - 18]:

$$A\ (Bq\ kg^{-1}) = \frac{(N_{Ei})_{net}}{\epsilon_{Ei}\ I_{Ei}\ t\ M} \quad (1)$$

where $(N_{Ei})_{net}$ = net photopeak area at energy (E) of radionuclide i (counts),

$(N_{Ei})_{net} = (N_{Ei})_S - (N_{Ei})_B$

A = specific activity concentration of individual radionuclide i for a peak at energy E in soil samples (Bq kg^{-1})

N_S = total net counts at energy E of radionuclide i (counts),

N_B = background net counts at energy E of radionuclide i (counts),

ϵ_{Ei} = the photopeak detection efficiency of a particular γ-ray energy (E),

I_{Ei} = absolute intensity corresponding to the photopeak at energy (E),

t = counting time of the soil sample (s)

M = mass of the measured soil sample (kg)

The activity concentration of ^{238}U was determined by measuring the average activity of γ-ray lines of its daughter products: ^{214}Pb (77.11 keV) and ^{234}Th (92.83 keV), while the activity concentration of ^{232}Th was determined by evaluating the average activity of γ-ray lines of its daughter products: ^{228}Ac (964.77 and 1630.63 keV). The γ-ray energy peak at 1460.8 keV (10.8%) of ^{40}K was used directly to determine the activity concentration of ^{40}K.

RADIATION HAZARD

Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity (Ra_{eq}) has been used for the assessment of radiological hazard of radioactivity in environmental materials. The exposure due to the γ- rays of materials that contain ^{238}U, ^{232}Th and ^{40}K is defined in terms of the radium equivalent activity Ra_{eq} is given by [UNSCEAR−2000, 17, 19]:

$$Ra_{eq}\ (Bq\ kg^{-1}) = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \leq 370\ (Bq\ kg^{-1}) \quad (2)$$

where A_{Ra}, A_{Th} and A_K are specific activity concentration of radium, thorium and potassium respectively. It has been assumed in this work that ^{226}Ra is in equilibrium with ^{238}U. The upper limit of radium equivalent activity is 370 Bq kg^{-1} [UNSCEAR- 1988, UNSCEAR- 2000].

Absorbed Dose Rate in Air (D)

The absorbed dose rate in air outdoors (nGy h^{-1}) at 1m above the ground surface due to the specific activity concentrations of ^{226}Ra, ^{232}Th series and ^{40}K is defined by [UNSCEAR−1988, 24]:

$$D\ (nGy\ h^{-1}) = 0.427\ A_{Ra} + 0.662\ A_{Th} + 0.043\ A_K \quad (3)$$

Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent due to the activity in the soil and building materials is calculated using the following equation [UNSCEAR-2000]:

$$AEDE\ (mSv\ y^{-1}) = D\ (nGy\ h^{-1}) \times 8760\ (h\ y^{-1}) \times 0.2 \times 0.7\ (Sv\ Gy^{-1}) \times 10^{-6} \quad (4)$$

where the value 0.7 Sv Gy^{-1} refers to the conversion coefficient from absorbed dose in air to effective dose received by adults, 8760 hours refers to the time in hours in one year, 0.2 represent the outdoor occupancy factor [UNSCEAR−2000] and D is the absorbed dose rate given in Eq. (3). The maximum permissible limit of Ra_{eq} activity is 370 $Bqkg^{-1}$ corresponds to annual effective dose equivalent of 1 mSv y^{-1} for general public (ICRP-1990).

External (H_{ex}) and Internal (H_{in}) Radiation Hazard Indices

The external radiation hazard index (H_{ex}) is defined by [20]:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

The value of external radiation hazard index must be less than 1 or equal to 1 [19]. The index value of 1 is equivalent to the upper limit of radium equivalent activity of 370 Bq kg^{-1} [19].

Internal radiation hazard Index (H_{in}) is defined by [19]:

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

H_{in} should be less than 1 for the radiation hazard to be negligible.

RESULTS AND DISCUSSIONS

Table 1 represents the measured ranges and arithmetic mean specific activity concentration values ± standard deviation (S.D.) in Bq kg^{-1} for ^{238}U, ^{232}Th series and ^{40}K in surface soil samples collected from nine different locations across old phosphate mine compared with the worldwide median values (WMVs) reported by UNSCEAR− 2000. The ranges of specific activity concentrations (SACs) of ^{238}U and ^{232}Th were found to be from 27.55 ± 7.55 to 420.19 ± 60.25 and from 222.35 ± 8.15 to 366.68 ± 92.47 Bq kg^{-1}, with an average values of 207.10 ± 21.02 and 265.95 ± 30.84 Bq kg^{-1}, respectively, in the surface soil samples collected from old phosphate mine.

Table 1: Specific Activity Concentrations of ^{238}U, ^{232}Th Series and ^{40}K in Surface Soil of Old Phosphate Mine in Russaifa of Jordan

Sample Location	Specific activity concentrations (Bq kg^{-1})		
	^{238}U ± S.D.	^{232}Th ± S.D.	^{40}K ± S.D.
1	27.55 ± 7.55	253.01 ± 20.55	2.40 ± 0.20
2	29.58 ± 0.92	239.02 ± 13.72	1.10 ± 0.10
3	50.20 ± 10.10	222.35 ± 8.15	0.90 ± 0
4	206.88 ± 3.48	366.68 ± 92.47	1.28 ± 0.225
5	321.85 ± 31.40	340.95 ± 87.76	BDL
6	195.46 ± 3.78	208.93 ± 45.61	1.25 ± 0.25
7	267.07 ± 0.56	225.54 ± 11.64	0.70 ± 0.02
8	345.15 ± 46.85	237.10 ± 21.54	0.43 ± 0.23
9	420.19 ± 60.25	300.02 ± 19.08	BDL
Mean±S.D.(This work)	207.10 ± 21.02	265.95 ± 30.84	0.895 ± 0.105
Range±S.D. (This work)	27.55 ± 7.55 − 420.19 ± 60.25	222.35 ± 8.15 − 366.68 ± 92.47	BDL −2.40 ± 0.20
Worldwide Median Value*	35	30	400
Worldwide Range*	17 −60	11 −64	140 −850

BDL = Below detection limit
* = UNSCEAR−2000

The measured concentration values of ^{238}U and ^{232}Th in surface soil of old phosphate mine in Russaifa were found to be higher than the WMVs [UNSCEAR-2000] due to the phosphate rocks are highly radioactive that contain trace amounts of ^{238}U and ^{232}Th series [UNSCEAR-1988]. This is in agreement with the data published by Al-Jundi (2002); Tufail et al., (2006); Al-Hammarneh and Awadallah (2009). The specific activity concentrations of ^{40}K were found to be lower than the concentration of both ^{238}U and ^{232}Th in all studied soil locations and world median values and its levels ranged from BDL to 2.40 ± 0.20 Bq kg^{-1} with mean value of 0.895 ± 0.105 Bq kg^{-1}.

Table 2 shows the range and mean values (with their standard deviation) of radium equivalent activity (Ra_{eq}), absorbed dose rate in air outdoors (D), annual effective dose equivalent (AEDE), external hazard index (H_{ex}) and internal hazard index (H_{in}) of ^{238}U, ^{232}Th series and ^{40}K in surface soils of old phosphate mine compared with the permissible limits

recommended by UNSCEAR- 1988, UNSCEAR- 2000 and ICRP-1990.

Table 2: The Experimental Results of Radium Equivalent activity (Ra_{eq}), Absorbed dose rate (D), Annual Effective dose Equivalent (AEDE), External (H_{ex}) and Internal (H_{in}) Radiation Hazard Indices due to the activity levels of γ-rays Emitted from ^{238}U, ^{232}Th Series and ^{40}K in all Surface Soil Samples of Old Phosphate Mine in Russaifa of Jordan Compared with the Recommended Limits by UNSCEAR-1988, UNSCEAR-2000 and ICRP-1990

Sample Location	Ra_{eq} (Bq kg^{-1})	D (nGy h^{-1})	AEDE (mSv y^{-1})	H_{ex}	H_{in}
1	389.53	179.35	0.219	1.052	1.126
2	371.45	170.91	0.209	1.003	1.083
3	368.23	168.67	0.207	0.994	1.130
4	731.31	331.13	0.406	1.975	2.534
5	809.42	363.14	0.445	2.186	3.056
6	494.32	221.82	0.272	1.335	1.863
7	589.64	263.37	0.323	1.592	2.314
8	684.23	304.35	0.373	1.848	2.782
9	849.20	378.03	0.464	2.290	3.430
Mean±S.D.(This work)	587.49 ± 179.6	264.53 ± 78.6	0.32 ±0.09	1.58 ± 0.48	2.14 ± 0.84
Range (This work)	368.23 −849.2	168.67 −378.03	0.207 −0.464	0.994 −2.29	1.083 −3.43
World Median Value*	< 370	57	0.48	≤ 1	≤ 1
World Range*		18 – 93	0.3 – 0.6		

* = UNSCEAR−2000

The estimated radium equivalent activity of surface soils vary from 368.23 to 849.2 Bq kg^{-1} with an average value of 587.49 ± 179.6 Bq kg^{-1} were exceeds the permissible level of 370 Bq kg^{-1} recommended by UNSCEAR-2000. The absorbed dose rates were calculated for each sample at one meter above ground surface in air outdoors using Eq. (3). Gamma absorbed dose rates lies between 168.67 and 378.03 nGy h^{-1} with an average value of 264.53 ± 78.6 nGy h^{-1}. This indicates that the absorbed dose rate from terrestrial radiation in air outdoors in the investigated area were higher than the permissible limit recommended by UNSCEAR-2000 and ICRP - 1990. This is expected since the phosphate mine at Russaifa has high level of uranium and thorium [UNSCEAR−1988, 14, 24]. The annual effective dose equivalent due to the specific activity levels of ^{226}Ra, ^{232}Th and ^{40}K were calculated according to Eq. (4) were found to be vary from 0.207 to 0.464 mSv y^{-1} with mean value of 0.32 ± 0.09 mSv y^{-1}. The range and average values for the annual effective dose rate were slightly less than the worldwide median value recommended by ICPR 60 – 1990, UNSCEAR− 1988 and UNSCEAR−2000. The external and internal radiation hazard indices were calculated to be in the range of 0.994−2.29 with an average value of 1.58 ± 0.48 and in the range of 1.083−3.43 with an average value of 2.14 ± 0.84, respectively. Radiation hazard indices seen to exceed the permissible limits of 1.

CONCLUSIONS

The main goal of this study was to determine the concentration levels of naturally occurring radioactive materials in soil samples collected from nine different locations across the old phosphate mine at Russaifa of Jordan by means of gamma-ray spectroscopy using a hyper-pure germanium (HPGe) detector in a low background configuration. The measured values of specific activity concentrations and radium equivalent activities of ^{238}U, ^{232}Th series and ^{40}K in surface soils of old phosphate mine were found to be higher than the worldwide median values recommended by UNSCEAR-1988 and UNSCEAR-2000. This is expected since the phosphate mine at Russaifa has higher levels of uranium and thorium series [UNSCEAR−1988]. The radiological hazard to humans due to the radionuclides of ^{238}U, ^{232}Th series and ^{40}K in surface soils collected from the areas studied were assessed. Gamma ray absorbed dose rates at one meter

above ground surface in air outdoors in the investigated area were higher than the permissible limit recommended by UNSCEAR-2000 and ICRP-1990. The external and internal radiation hazard indices were calculated to be in the range of 0.994−2.29 with an average value of 1.58 ± 0.48 and in the range of 1.083 −3.43 with an average value of 2.14 ± 0.84, respectively were exceed the permissible limits of 1 [ICRP – 1990].

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